Title

Intra-aggregate biogeochemical dynamics of chromium contamination and in-situ remediation

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Intra-aggregate Biogeochemical Dynamics of Chromium Contamination and In-situ Remediation

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Transport of redox-sensitive contaminants through the vadose zone is typically complex because of the broad range of transport times and reaction rates encountered over short distances. Multi-region flow and transport models are often used to describe fast advective transport through fractures and macropores, and slower diffusion-dominated transport within sediment blocks and soil aggregates. However, transport and reactions occurring within the diffusion-controlled domains that often make up most of the subsurface are commonly only inferred or assumed. Direct measurements within soil aggregates and sediment blocks are needed to understand biogeochemical processes. This is demonstrated through laboratory studies of chromium contamination of soil aggregates, and subsequent in-situ remediation (reduction of Cr(VI) to Cr(III)) by organic carbon infusion. Spatially resolved determination of Cr concentrations and oxidation states using micro-XANES, and spatially-resolved microbial community analyses were done on synthetic and natural soil aggregates. During the diffusion-limited contamination process, more Cr(VI) was transported, but to shorter distances, in more microbially active aggregates. Sharply terminated diffusion fronts, within 2 to 10 mm of the aggregate surface, result from increasing Cr(VI) reduction rates with depth. Infusion of organic carbon into previous Cr(VI)-contaminated aggregates resulted in more rapid reduction to Cr(III) with higher organic carbon concentrations, and lower reduction rates in more highly contaminated sediments. These results show that intra-aggregate Cr dynamics are strongly diffusion-limited in more microbially active systems, and that bulk soil chemical and microbial characterization can obscure relevant biogeochemical processes.

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